Supplementary Information

Efficiency enhancement of planar perovskite solar cells by adding zwitterion/LiF double interlayers for electron collection

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Materials and chemicals

Patterned indium tin oxide (ITO) glass substrates (10 Ω/□) were supplied by NSG group. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Clevios P VP Al 4083) was purchased from Heraeus Holding GmbH. Methylammonium iodide (MAI) was obtained from Dyesol Ltd and 1-Material Inc. [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) was bought from Nano-C Inc. Other materials, includinglead (II) iodide (PbI_2, 99% purity), dimethyl sulfoxide (DMSO, anhydrous, ≥99.9% purity), γ -butyrolactone (GBL, ≥99% purity), rhodamine 101 inner salt, LiF (≥99% purity), chlorobenzene (anhydrous, 99.8% purity) and isopropanol (IPA, anhydrous, 99.5% purity),were supplied by Sigma-Aldrich. All materials are used without further purification.

Thin film characterizations

X-ray diffraction (XRD) patterns were acquired using a Bruker D8 Advance XRD Instrument. Absorption spectra were recorded on a Shimadzu UV-1800 spectrophotometer. Electron scanning microscopic (SEM) images were obtained with a Philips XL 30 SEM. The cross-sectional SEM images were obtained using a Zeiss Supra-40 SEM with in-lens detector.Atomic force microscopic (AFM) images were acquired usinga VeecoNanoScope IV Multi-Mode AFM operated in tappingmode. Film thickness was determined by a surface profilometer (KLA Tencor, Alpha-Step IQ).

Device fabrication and characterizations

Planar perovskite solar cells (PSCs) were fabricated on pre-patterned ITO glass substrates. The ITO glass substrates were sequentially cleaned with detergent, de-ionized water, acetone and isopropyl alcohol. They were cleaned in a UV ozone oven for 15 min before the device fabrication.A thin layer (*ca.*30 nm thick) of PEDOT:PSS was spin coated on ITO substrates at 6000 rpm for 1 min. It wasthen baked on a hot plate at 140 °C for 10 min. The substrates were transferred into a glove box filled with dried nitrogen (O_2≤ 1 ppm; H_2O≤ 1 ppm).A perovskite photoactive layer was deposited by spin coating a solution containing 0.14M (39 mg) $PbCl_2$, 1.26M (581 mg) PbI_2 and 1.3M (209 mg)

MAI in 1 ml cosolvent of DMSO:GBL (3:7 vol. ratio). Both the perovskite precursor solution and substrates were pre-heated at 80℃ before spin coating. The spin coating was programmed to run at 1000 rpm for 20 s and then 4000 rpm for 60 s. At 40 s after the start of spin coating process, 160 ul of anhydrous toluene was injected onto the spinning film to quench it. The yellowish and transparent as-cast films were subsequently annealed at 100 ℃ for 20 min. The thickness of the perovskite layer was *ca.*260 nm as determined by the surface profilometer. A 50 nm-thick layer of PCBM was deposited on the perovskite layer by spin coating (20 mg/ml in chlorobenzene, 1500 rpm 40s).For some cells, 0.5 mg/ml rhodamine 101/IPA was spin cast at 2000 rpm onto the PCBM layer. The films were then transferred to a metal evaporation chamber, where LiF (1 nm) and Ag (100 nm) were deposited through a shadow mask (active area was 0.11 cm^2) at approximately 1 x 10^{-6} mbar. The complete solar cells were transferred back to the glove box and encapsulated with UV-curable epoxy and cover glass slides before testing in air.

The photovoltaic performance of the PSCs was measured with a computer-programmed Keithley 2400 source/meter under a Newport'sOriel class A solar simulator, which simulated the AM1.5sunlight with energy density of 100 mW cm^{-2} and was certified to theJIS C 8912 standard. The active area of each device was 0.11 cm^2.Incident photon-to-current conversion efficiencies (IPCEs) of PSCs were measured with a 300W Xenon Lamp (Oriel 6258) and a Cornerstone 260 Oriel 74125 monochromator.

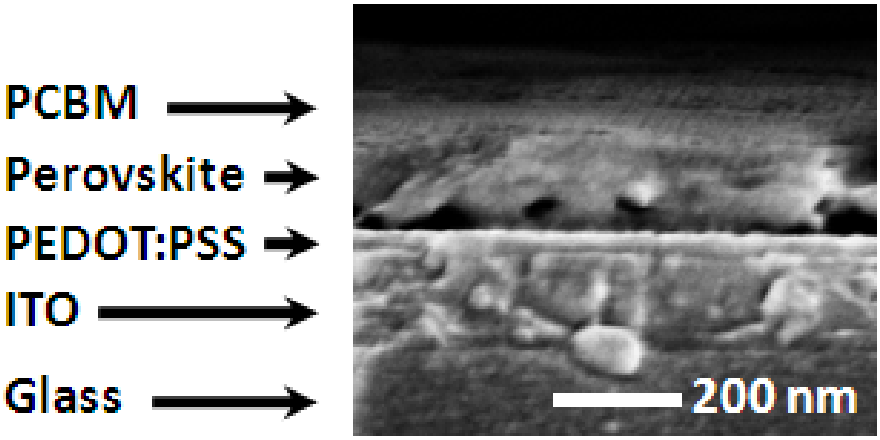

Figure S1. Cross-sectional SEM image showing the cell architecture of a planar perovskite solar cell without the electron-collection interlayer and the top Ag electrode.

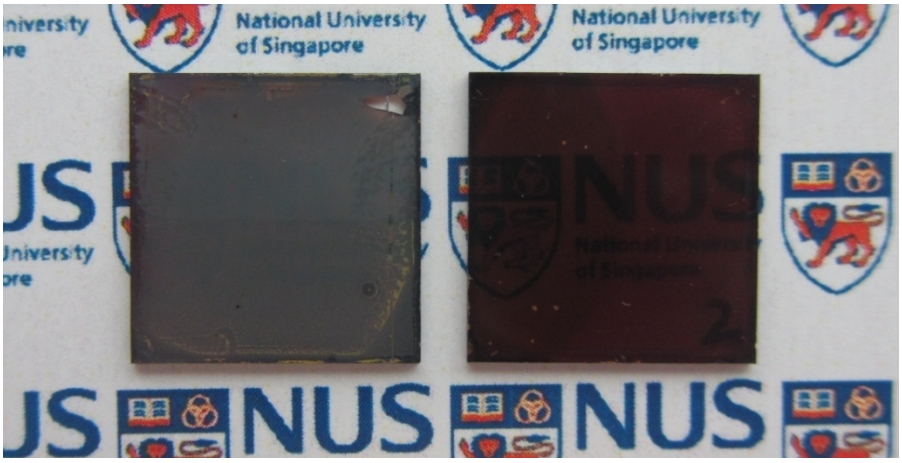

Figure S2. Photos of two perovskite films without (left) and with (right) a layer of PCBM on top. The photos were taken after two films were stored in air (*ca.* 25 °C and 90 % humidity) for one day.

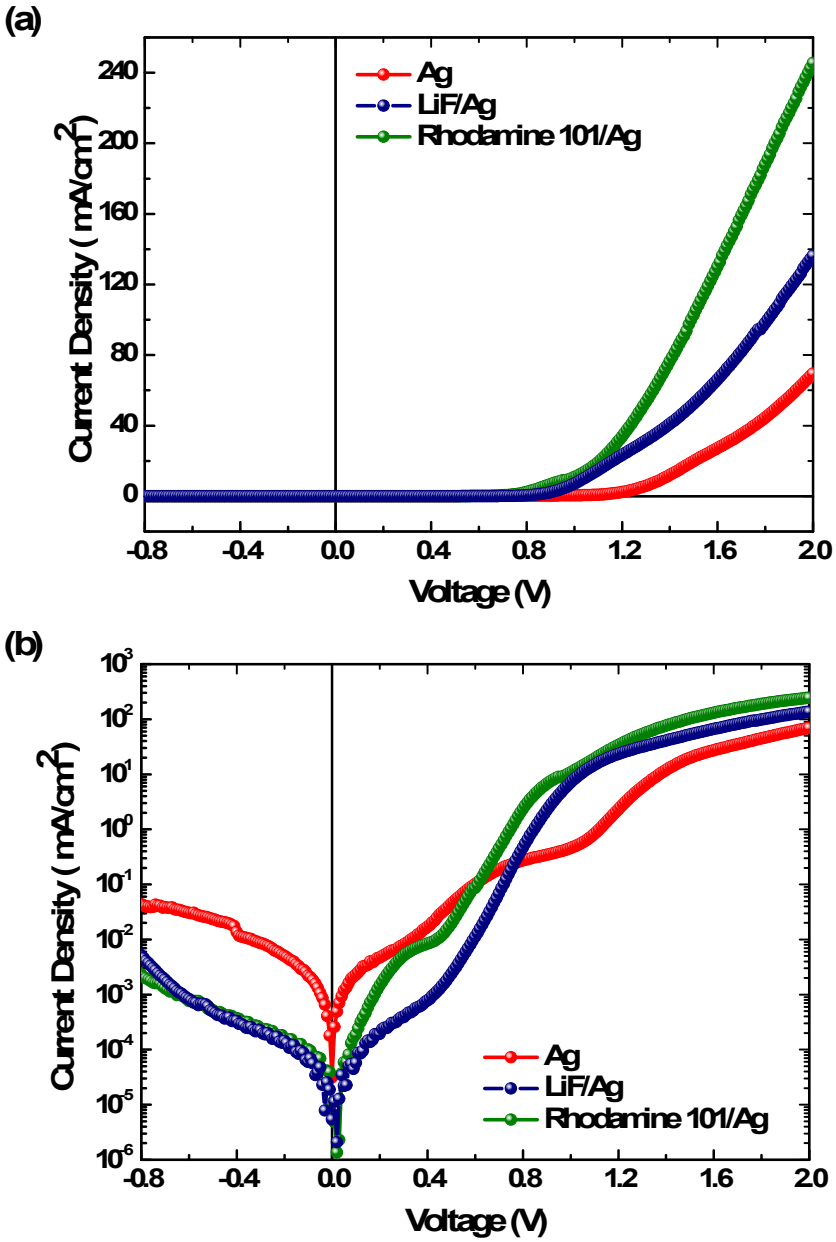

Figure S3. *J-V* curves of perovskite solar cells with different cathode configurations in dark.The data in (a) are present in linear scale, and the same data are presented in (b) in semi-log scale.